Spallation with High Energy Protons as a Source of ^{178m2}Hf isomers: Optimization and Accumulation

1. Introduction

The production of ^{178m2}Hf isomers continues to be a major task of the research work related with the use of these isomers for gamma ray lasers development. Recent results obtained by the research group from the Center for Quantum Electronics of UT Dallas (CQE) [1,2] from irradiations with low energy photons of isomeric samples at Spring8, made the question of how can we produce ^{178m2}Hf isomers in macroscopic quantities more actual than ever. The new results showing the possibility to trigger the release of the energy stored in the high spin ^{178m2}Hf isomers through irradiation of low energy photons, represent a strong confirmation of the previous reports [3,4,5] since it made use of a sophisticated accelerator for x-ray photon production where the incident photon flux and the transmitted one through the irradiated samples as well as the photons energy were well monitored and controlled. The acquisition and analysis of the signals from the spectroscopic amplifier were performed with a high-tech sampling system developed at CQE [6,7] that allowed an accurate measurement of the gamma-ray energy spectra.

The main problem that we have still to face is the availability of the isomeric material. The existing inventory is continues to diminish being using in different laboratories for test irradiations while there not yet developed a method for producing large quantities of ^{178m2}Hf isomers. The IGE Foundation in Bucharest has contracted with EOARD contracts for the investigation of spallation with high-energy protons as a source for isomers production. The data available in the literature on these processes was fragmentary and incomplete so that we started a systematic study of the spallation reaction that can lead to

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the production of ^{178m2}Hf isomers. In 2000 we have reported on the spallation of Ta samples with protons of different energies and concluded that the process can be optimized for a higher yield of isomers, better isomer-to-ground state ratio, less contamination, shorted cooling times, by choosing the right irradiation energy, target thickness, irradiation period.

After this round of measurements naturally occurred the question whether the irradiation of other materials with high-energy protons would be better for the isomer production. Two options were available: natural Re and ^{186}W. Since natural Re was available immediately in large quantities at low cost we decided to start with it. The results obtained from the irradiation of natural Re samples with protons at different energies make the object of this report.

2. Experiment and Results

For the study of the spallation of natural Re with high-energy protons the IGE Foundation has contracted with a group of scientists at JINR Dubna for a 50 hrs. irradiation of samples at the LNP synchrocyclotron.

The irradiated samples were made of natural Rhenium that consists of only two isotopes: 185 – 37.4% and 187 – 62.6% and has a specific weight of 21.02 g/cm^2. Four samples had a thickness of about 21 g/cm^2 (~ 10 mm thick) while a fifth one had 0.556 g/cm^2 (~ 0.26 mm thick). The four thick samples were irradiated at four proton energies, 150, 300, 450 and 660 MeV, respectively. The thinner one was irradiated together with an Al foil of similar thickness at 660 MeV for absolute cross section calibration purposes. After three weeks of cooling the samples their gamma activity was repeatedly measured with Ge detectors. Due to the cooling time isotopes that have half-lives of less than 2 days could not be identified. These measurements gave the cross sections for those isotopes produced with higher yield. Weak products as ^{178m2}Hf were covered by the strong activity of the other products and chemical processing of the samples was needed to separate only the Hf fraction and to measure accurately how many isomers were produced. This

method allowed identifying isotopes produced with cross sections as low as 1 μbarn. The statistical error on the results is of the order of 15%.

Tables 1 and 2 summarize the results of the measurements at the four proton energies.

Table 1. Yields of isotopes and mean cross-sections measured after activation of the ^{nat}Re target of a 21g/cm^2 thickness with 660 MeV protons. Statistical inaccuracy is typically on a level of 15%. The theoretically predicted yield values are given in the last column.

Nuclide	$T_{1/2}$	E_γ, keV	Type of yield	Mean σ, mbarn	Yield, atoms/proton	Predicted yield
^{185}Os	93.6 d	646.1	Indep.	4.4	$3.0 \cdot 10^{-4}$	$4.0 \cdot 10^{-4}$
^{184g}Re	38 d	903.3	Indep.	38	$2.58 \cdot 10^{-3}$	$\} 3.5 \cdot 10^{-3}$
^{184m}Re	165 d	920.9	Indep.	11	$7.5 \cdot 10^{-4}$	
^{183}Re	70 d	162.5	EC cum.	62	$4.22 \cdot 10^{-3}$	$3.3 \cdot 10^{-3}$
^{182g}Re	2.67 d	1427.3	Indep.	17	$1.16 \cdot 10^{-3}$	$2 \cdot 10^{-3}$ *)
^{178}W	21.7 d	1340.9	EC cum.	36	$2.45 \cdot 10^{-3}$	$3.7 \cdot 10^{-3}$
^{182}Ta	115 d	1231.0	Indep.	2.3	$1.57 \cdot 10^{-4}$	$2.3 \cdot 10^{-4}$
^{181}Hf	42.6 d	482.0	β^- cum.	0.14	$0.95 \cdot 10^{-5}$	$2.1 \cdot 10^{-5}$
^{179m2}Hf	25.1 d	453.7	Indep.	0.12	$0.82 \cdot 10^{-5}$	$4.2 \cdot 10^{-5}$ *)
^{178m2}Hf	31 y	574.2	Indep.	0.13	$0.9 \cdot 10^{-5}$	$7.3 \cdot 10^{-5}$ *)
^{175}Hf	70 d	343.4	EC cum.	59	$4.01 \cdot 10^{-3}$	$4.1 \cdot 10^{-3}$
^{172}Hf	1.87 y	1093.6	EC cum.	55	$3.74 \cdot 10^{-3}$	$3.9 \cdot 10^{-3}$
^{177m}Lu	160.9 d	418.5	Indep.	0.034	$2.3 \cdot 10^{-6}$	$8 \cdot 10^{-6}$ *)
^{174g}Lu	3.31 y	1241.8	Indep.	0.50	$3.4 \cdot 10^{-5}$	$\} 5.2 \cdot 10^{-5}$
^{174m}Lu	142 d	992.0	Indep.	0.59	$4 \cdot 10^{-5}$	
^{173}Lu	1.37 y	272.2	EC cum.	61	$4.15 \cdot 10^{-3}$	$4.15 \cdot 10^{-3}$
^{171}Lu	8.22 d	739.8	EC cum.	46	$3.13 \cdot 10^{-3}$	$4.0 \cdot 10^{-3}$
^{170}Lu	2 d	985.1	EC cum.	42	$2.86 \cdot 10^{-3}$	$3.8 \cdot 10^{-3}$
^{169}Yb	32 d	307.7	EC cum.	57	$3.88 \cdot 10^{-3}$	$3.7 \cdot 10^{-3}$
^{166}Yb	2.36 d	1374.1	EC cum.	39	$2.65 \cdot 10^{-3}$	$3.6 \cdot 10^{-3}$
^{168}Tm	93.1 d	720.3	Indep.	0.30	$2.0 \cdot 10^{-5}$	$1.5 \cdot 10^{-5}$
^{167}Tm	9.24 d	531.5	EC cum.	50	$3.40 \cdot 10^{-3}$	$3.3 \cdot 10^{-3}$
^{166}Dy	3.4 d	1379.6	β^- cum.	≤ 2	$\leq 1.4 \cdot 10^{-4}$	-
^{160}Tb	72.3 d	1271.9	Indep.	0.17	$1.2 \cdot 10^{-5}$	$4.2 \cdot 10^{-7}$
^{156}Tb	5.35 d	534.3	Indep.	0.83	$5.6 \cdot 10^{-5}$	$2 \cdot 10^{-5}$
^{155}Tb	5.32 d	367.4	EC cum.	13.4	$0.91 \cdot 10^{-3}$	$1.5 \cdot 10^{-3}$

^{153}Gd	242 d	103.2	EC cum.	11.5	$0.78 \cdot 10^{-3}$	$1.1 \cdot 10^{-3}$
^{151}Gd	120 d	243.2	EC cum.	8.0	$5.4 \cdot 10^{-4}$	$8.5 \cdot 10^{-4}$
^{149}Gd	9.4 d	298.5	EC cum.	7.4	$5.0 \cdot 10^{-4}$	$5.5 \cdot 10^{-4}$
^{146}Gd	48.3 d	633.7	EC cum.	4.5	$3.1 \cdot 10^{-4}$	$4.8 \cdot 10^{-4}$
^{156}Eu	15.2 d	2097.7	β^- cum.	0.19	$1.3 \cdot 10^{-5}$	-
^{149}Eu	93 d	277.0	EC cum.	8.2	$5.6 \cdot 10^{-4}$	$6 \cdot 10^{-4}$
^{148}Eu	54.5 d	550.3	Indep.	0.26	$1.8 \cdot 10^{-5}$	$7 \cdot 10^{-5}$
^{147}Eu	24.0 d	601.4	EC cum.	4.5	$3.1 \cdot 10^{-4}$	$6 \cdot 10^{-4}$
^{145}Eu	5.94 d	893.7	EC cum.	1.9	$1.3 \cdot 10^{-4}$	$4 \cdot 10^{-4}$
^{148g}Pm	5.37 d	914.9	Indep.	0.09	$0.6 \cdot 10^{-5}$	} $4.2 \cdot 10^{-7}$
^{148m}Pm	41.3 d	286.6	Indep.	0.03	$2 \cdot 10^{-6}$	
^{144}Pm	363 d	476.8	Indep.	0.06	$4 \cdot 10^{-6}$	$2.3 \cdot 10^{-5}$
^{143}Pm	265 d	742.0	EC cum.	1.6	$1.1 \cdot 10^{-4}$	$2.8 \cdot 10^{-4}$
^{139}Ce	137.7 d	165.8	EC cum.	0.87	$5.9 \cdot 10^{-5}$	-
^{140}Ba	12.8 d	1596.5	β^- cum.	0.006	$0.4 \cdot 10^{-6}$	-
^{131}Ba	11.8 d	496.3	EC cum.	0.07	$5 \cdot 10^{-6}$	$1.5 \cdot 10^{-5}$
^{121m}Te	154 d	212.3	Indep.	0.15	$1.0 \cdot 10^{-5}$	$0.8 \cdot 10^{-5}$ *)
^{113}Sn	115 d	391.7	EC cum.	0.11	$7.5 \cdot 10^{-6}$	
^{110m}Ag	250 d	884.7	Indep.	0.06	$4 \cdot 10^{-6}$	
^{106m}Ag	8.46 d	1045.8	Indep.	0.1	$7 \cdot 10^{-6}$	
^{105}Ag	41.3 d	443.4	EC cum.	0.13	$9 \cdot 10^{-6}$	
^{103}Ru	39.3 d	497.1	β^- cum.	0.14	$0.95 \cdot 10^{-5}$	
^{100}Pd	3.6 d	2376.1	β^+ cum.	0.01	$0.7 \cdot 10^{-6}$	
^{95}Nb	35 d	765.8	Indep.	0.21	$1.4 \cdot 10^{-5}$	
^{91m}Nb	62 d	1204.8	Indep.	0.018	$1.2 \cdot 10^{-6}$	
^{95}Zr	64 d	756.7	β^- cum.	0.10	$0.7 \cdot 10^{-5}$	
^{88}Zr	83.4 d	392.9	EC cum.	0.27	$1.8 \cdot 10^{-5}$	
^{88}Y	106.6 d	898.0	Indep.	0.49	$3.3 \cdot 10^{-5}$	
^{85}Sr	64.8 d	514.0	β^+ cum.	0.87	$5.9 \cdot 10^{-5}$	
^{84}Rb	32.9 d	881.6	Indep.	0.48	$3.3 \cdot 10^{-5}$	
^{83}Rb	86.2 d	520.4	β^+ cum.	0.75	$5.1 \cdot 10^{-5}$	
^{75}Se	119 d	264.6	β^+ cum.	0.55	$3.7 \cdot 10^{-5}$	
^{72}Se	8.4 d	834.0	β^+ cum.	0.2	$1.3 \cdot 10^{-5}$	
^{74}As	17.8 d	595.8	Indep.	0.55	$3.7 \cdot 10^{-5}$	
^{56}Co	78.8 d	1771.4	EC cum.	0.03	$2 \cdot 10^{-6}$	
^{59}Fe	44.5 d	1099.3	β^- cum.	0.24	$1.6 \cdot 10^{-5}$	
^{52}Mn	5.6 d	1434.1	β^+ cum.	0.022	$1.5 \cdot 10^{-6}$	
^{48}V	16 d	983.5	EC cum.	0.026	$1.8 \cdot 10^{-6}$	

*) *The calculated values correspond to the sum of the independent yields for both the isomeric and the ground states.*

Table 2.   Comparison of the measured and predicted values of mean cross-section (mbarn) for the isotopes detected after spallation of the ^{nat}Re target by protons with an incident energy of 450, 300 and 150 MeV.

Nuclide	450 MeV		300 MeV		150 MeV	
	exp.	theor.	exp.	theor.	exp.	theor.
^{185}Os	5.5	9.1	9.4	15	23	25
^{184g}Re	45	}55	49	}62	76	}63
^{184m}Re	13		14		24	
^{183}Re	82	61	100	81	140	104
^{178}W	55	76	82	102	136	145
^{182}Ta	2.3	2.3	1.9	1.3	1.1	0.34
^{181}Hf	0.14	0.15	0.085	0.071	0.015	0.012
^{179m2}Hf	0.13	0.43[*)]	0.08	0.14[*)]	0.02	0.018[*)]
^{178m2}Hf	0.1	0.63[*)]	0.09	0.27[*)]	0.03	0.035[*)]
^{175}Hf	93	80	96	91	58	50
^{172}Hf	64	68	63	66	12	11
^{177m}Lu	0.03	0.09[*)]	0.024	0.022[*)]	0.004	-
^{174g}Lu	0.50	}0.48	0.31	}0.26	0.045	}0.035
^{174m}Lu	0.68		0.47		0.07	
^{173}Lu	89	74	91	74	18	18
^{171}Lu	81	66	78	58	5.1	5.7
^{170}Lu	68	59	53	48	-	2.6
^{169}Yb	78	54	43	39	0.66	1.14
^{166}Yb	47	45	27	23	-	0.05
^{168}Tm	0.21	0.12	0.12	0.07	-	-
^{167}Tm	56	43	26	25	-	0.14
^{156}Tb	0.24	0.09	-	0.006		
^{155}Tb	8.0	6.5	-	0.28		
^{151}Gd	1.8	2.1	-	0.03		
^{149}Gd	1.5	1.3	0.18	0.007		
^{146}Gd	0.66	0.65	0.04	0.001		
^{156}Eu	0.03	-	0.07	-		
^{149}Eu	1.64	1.5	-	0.01		
^{148}Eu	0.053	0.13	0.02	-		
^{147}Eu	0.66	0.88	-	0.0007		
^{145}Eu	0.4	0.46		-		
^{148g}Pm	0.016	}0.003				
^{148m}Pm	0.008					
^{144}Pm	0.011	0.018	-	0.003		
^{143}Pm	0.26	0.17	-	0.006		
^{140}Ba	0.0024					
^{110m}Ag	0.077		0.07			

^{106m}Ag	0.17		0.05			
^{105}Ag	0.24		-		0.014	
^{103}Ru	0.10		0.12		0.08	
^{95}Nb	0.20		0.17		0.058	
^{91m}Nb	0.29		0.25			
^{95}Zr	0.14		0.12		0.040	
^{88}Zr	0.40		0.37		0.012	
^{88}Y	0.34		0.22		0.043	
^{85}Sr	0.69		0.54		-	
^{84}Rb	0.45		0.31		-	
^{83}Rb	0.87		0.51		-	
^{75}Se	0.58		0.38		0.06	
^{72}Se	0.21		0.17		-	
^{74}As	0.45		0.22		-	
^{56}Co	0.03		0.02		0.01	
^{59}Fe	0.18		0.2		0.03	
^{52}Mn	0.016		-		-	
^{48}V	0.013		0.012		0.005	

$^{*)}$ *The calculated values correspond to the sum of the independent yields for both the isomeric and the ground states.*

3. Discussion

The experimental data are compared to the calculations performed with the computer code LAHET [8]. This code that makes use of the Monte Carlo technique and sophisticated transport approaches suites the best the calculation of the spallation processes at intermediate energies. For more details on the spallation process and the LAHET simulations see Ref. [9]. The results of the calculations are compared to the experimental ones in Tables 1 and 2.

At a first look to these tables we can notice the extension of the identified isotopes to much lower masses as compared with the Tantalum spallation. This is due to a higher probability for fission in the case of p + Re system. The fission products correspond to masses A < 120 while spallation to masses A > 135. In the region 120 < A < 135 both mechanisms contribute to the yield of the isotopes. The LAHET code cannot account for

fission and this is the reason why below mass A=120 we don't have anymore calculated yields.

In the Tables we have two types of yields: cumulative and independent. The cumulative yields correspond to isotopes that accumulate the total yield of the isobaric chain following electron capture (EC) and ß-decay processes. The independent yields correspond to isotope population directly from the reaction. One can see that the most abundant isotopes are due to cumulative yield.
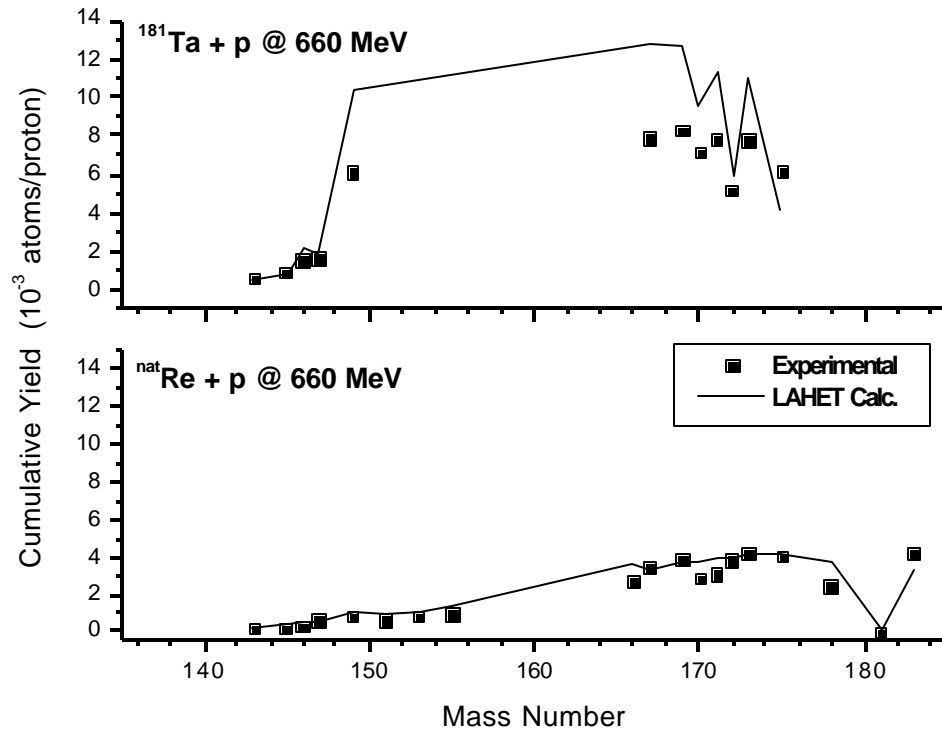

Figure 1. Comparison of the cumulative yields for the spallation with 660 MeV protons of Tantalum (upper panel) and natural Rhenium (lower panel) samples. Filled squares correspond to the measurement with a 15% of statistical error. The continuous lines correspond to the calculated yields.

Fig. 1 shows a comparison of the cumulative yields experimentally measured for the cases of spallation of Tantalum and Rhenium samples at proton energy of 660 MeV.

We can see that the computer calculations give a good description of the cumulative yields. In the same time, from the tables, we notice that for the isomers populated as independent yield directly from the reaction (with ground state that cannot be measured) the description is rather poor. This is the case of ^{178m2}Hf isomers. At 660 MeV the calculations predict a total yield of 7.3×10^{-5} atoms/proton while the measured value is 0.9×10^{-5} atoms/proton. This problem emerges from the fact that the simulation describes total yield for a given isotope rather than different yields for isomers and ground state. This is a limitation that we should address in the next future in order to get realistic predictions for the yields of isomers.

As seen in Fig. 1, in the case of the natural Re samples the maximum of the yield distribution has moved to higher masses around A = 178 in agreement with the judgment made before that spallation favors many nucleons evaporation channels. In both cases of spallation of Tantalum and Rhenium we can see that the maximum of yield distribution is located about 10 mass units away from the p + target system.

The contaminant background has two main consequences: it increases the radioactivity of the targets leading to long cooling times and it prevents the accurate measurement of the low activity due to the isomer decay (Compton background in the region of interest, high counting rates that implies the use of small solid angle for the measurements).

We divide the sources of background in two categories: the ones that cannot be eliminated from the samples through chemical separation and other types. In the first category we have ^{172}**Hf** with a half-life of **1.87 yrs**. and ^{175}**Hf** with a half-life of **70.4 days**. The ^{172}Hf nuclei through EC processes lead to ^{172}Yb nuclei in excited states that decay through many gamma rays towards its ground state. The cooling down of the ^{172}Hf activity is very lengthy. The ^{175}Hf nuclei decay through EC to excited states in ^{175}Lu nuclei that at their turn decay gamma to the ground state. The most intense gamma radiation is of 343 keV and the high counting rate produced by it could be reduced using Pd filters and varying the target-detector distance. However, due to its short half-live the ^{175}Hf contamination will disappear in a few years from the samples. **The only way to avoid these sources of background is to minimize their production.**

In the category others we included the isotopes that accumulate in the sample during the irradiation and which decay to stable ground states of the same species as the isomer (namely Hf isotopes). Their effect is to reduce the isomer-to-ground state ratio and to deteriorate the chemical composition of the target. This is the case of ^{178}**W** nuclei ($T_{1/2} =$ **21.6 days**) that through EC decays transform into ^{178}Hf nuclei in ground state. **In order to reduce the effects of these contaminants one should change periodically the targets and to chemically remove them from the samples.**
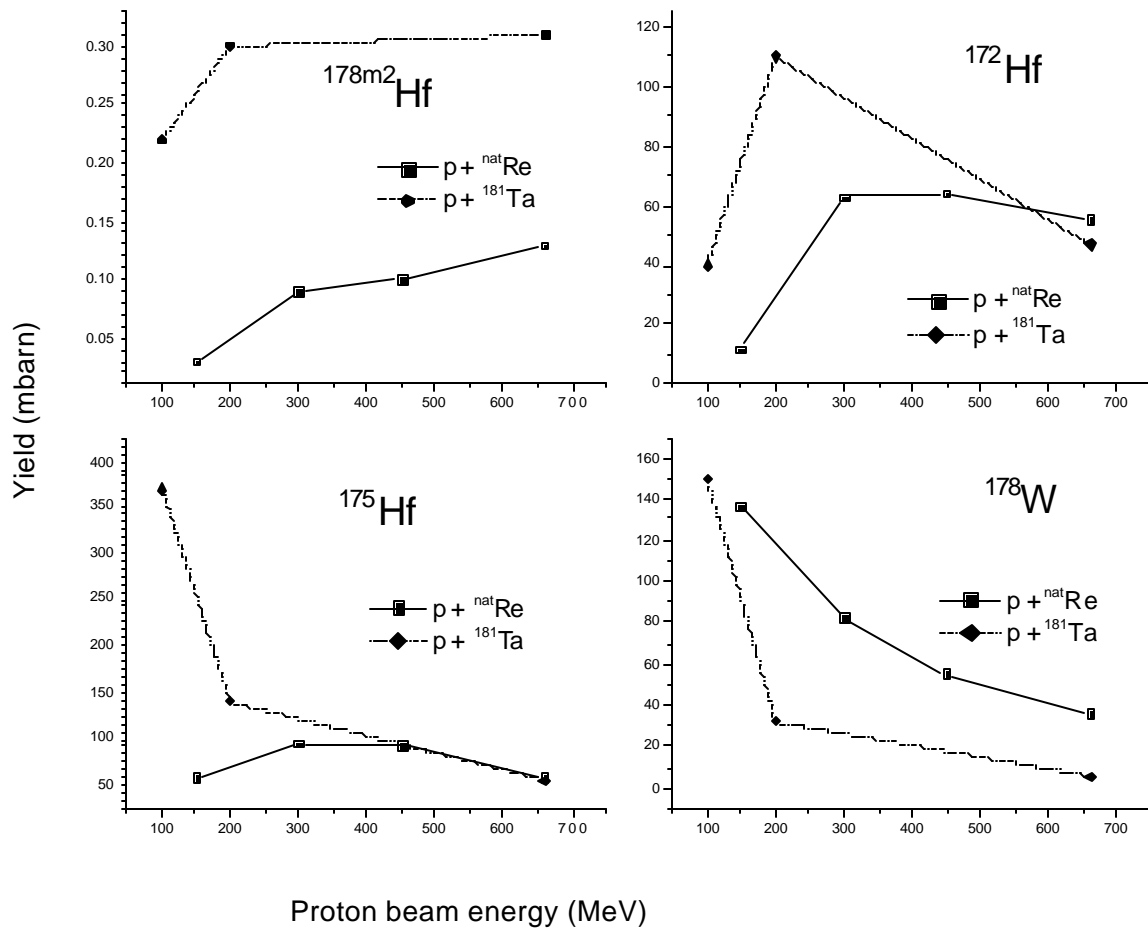

Figure 2. Measured production yields for ^{172m2}Hf and the main contaminants (^{172}Hf, ^{175}Hf and ^{178}W) compared for the spallation of ^{181}Ta and ^{nat}Re, respectively, with high-energy protons.

Fig. 2 shows a comparison of the measured production yields for the ^{178m2}Hf isomers and for the main contaminants produced through spallation of the ^{181}Ta and ^{nat}Re samples with high-energy protons. We notice that for the ^{178m2}Hf isomers production we get a factor 3 less in the case of ^{nat}Re spallation compared to the ^{181}Ta spallation. There is an improvement for production of ^{172}Hf (which is the most disturbing contaminant) at intermediate energies. **From the figure it results that higher energies favor a higher isomer production while the contaminant production has a decreasing trend (if we discard the lower energy point which is the most unfavorable for isomer production).**

The very low yield of the ^{178m2}Hf isomers is mainly due to the fact that Hf isotopes are far from the most probable charge populated by spallation. Spallation processes favor the population of neutron-deficient nuclei far from the ß-stability line that via EC or ß-decays ends in long-lived states of isobars situated closer to the stability line. **A way to overcome this problem is to use a target built of very neutron-rich nuclei. This is the case of ^{186}W. The use of this target should lead to an important increase of the yield of ^{178}Hf isotope**.

4. Conclusions

We report the results obtained from the spallation of ^{nat}Re samples with high-energy protons for the optimization of the ^{178m2}Hf isomers production. We performed irradiations at four different proton energies: 150, 300, 450 and 660 MeV. The experimental results are compared with Monte Carlo simulations performed with the computer code LAHET that give a good description of the yields for the most probable nuclei to be produced through spallation but it fails to describe the production of the isomers since it cannot distinguish between isomeric and ground states. Using higher mass samples the fission probability increases very much leading to the production of more radioisotopes but they can be easily removed by chemical processing. The use of ^{nat}Re did not reduce drastically

the contaminant production as compared to the case of ^{181}Ta spallation but it leads to a reduction of the ^{178m2}Hf production. A way to improve the isomer production is to use a sample built of very neutron-rich nuclei, as it is the case of ^{186}W.

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